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ORCID logoORCID: <https://orcid.org/0000-0002-0756-9764> (2021) Screen-
printed electrodes: transitioning the laboratory in-to-the field. Talanta Open,
3. p. 100032.

Downloaded from: <https://e-space.mmu.ac.uk/628041/>

Version: Published Version

Publisher: Elsevier

DOI: <https://doi.org/10.1016/j.talo.2021.100032>

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Screen-printed electrodes: Transitioning the laboratory in-to-the field

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ARTICLE INFO

Keywords:

Electroanalysis
Electrochemistry
Analytical techniques
Sensors
Screen-Printed Electrodes (SPEs)

ABSTRACT

This short article overviews the use of screen-printed electrodes (SPEs) in the field of electroanalysis and compares their application against traditional laboratory based analytical techniques. Electroanalysis coupled with SPEs can offer low-cost, precise, sensitive, rapid, quantitative information and laboratory equivalent results. The combined use of SPEs and electroanalysis reduces the need of sample transportation and preparation to a centralised laboratory allowing experimentalists to perform the measurements where they are needed the most. We first introduce the basic concepts and principles of analytical techniques to the reader, with particular attention to electroanalysis, and then discuss the application of SPEs to common analytical targets such as food, environmental, forensics, cancer biomarkers and pathogenic monitoring and sensing.

1. Introduction

Analytical chemistry applies a combination of classical, wet chemical and modern instrumental methods to separate and identify matter in both qualitative and quantitative analysis in forensic, environmental, industrial and medical applications. Instrumental analysis dominates the area of modern analytical chemistry where experimentalists tend to try improving or discovering new methods/analysis or their applications to new topics. Spectroscopy, mass spectrometry, thermal, separation, microscopic and electrochemical analysis are among the most common instrumental methods utilised by experimentalists. Atomic absorption spectroscopy (AAS), atomic emission spectroscopy (AES), ultraviolet-visible spectroscopy (UV-VIS), x-ray spectroscopy, fluorescence spectroscopy, infrared spectroscopy (IR), Raman spectroscopy and nuclear magnetic resonance spectroscopy (NMR) are some examples of spectrometric techniques. Mass spectrometry involves the use of magnetic and electric fields to ionize an analyte, and study its mass-to-charge ratio of ions and obtain elemental information. Calorimetric and thermogravimetric techniques are thermal analysis methods that measure the interaction between a given material and heat. Chromatography and electrophoresis are the most common separation methods. Microscopic techniques include optical, electron and scanning probe microscopy. In the case of electroanalytical techniques, charge, current and potential are measured within electrochemical cells; being voltammetric, potentiometric and coulometric methods the most common ones.

In comparison to current analytical laboratory techniques, electrochemical methods are an affordable and easy to use solution, yet capable of providing sensitive results with little equipment in short turnaround times. In this short literature review, we compare the use and perfor-

mance of screen-printed electrodes (SPEs) as the basis of electroanalytical sensing platforms against other traditional analytical techniques. Recent developments are summarised herein and we highlight future promising advances in this exciting topic.

1.1. Electroanalytical methods

Electrochemical methods, when compared to common laboratory equipment, offer the advantageous of easy sample preparation, little or no installation and multi-analyte detection at a low-cost to name a few. The main interest of electrochemical experiments is the solution-surface of the electrode interface [1–3]. Potentiostatic methods measure electroactivity by monitoring the potential difference between working (WE), counter (CE) and reference electrodes (RE) respectively. Amperometric methods measure the change in the oxidation state of the electroactive species (Faradaic current), which is directly proportional to the analyte's concentration. In comparison to more often utilised laboratory analytical techniques such as spectrophotometry, HPLC (high-performance liquid chromatography) or colorimetric kits that require bulky, complex to perform, time-consuming and expensive instrumentation, the electrochemical sensing of target analytes (termed electroanalysis) offers fast, precise, portable and affordable methods/devices whilst also offering high sensitivity and selectivity towards electroactive analytes [1, 3]. *Why are electroanalytical techniques so sensitive in comparison to other analytical approaches?* The answer lies in the Randles–Ševčík equation. Let's consider the Randles–Ševčík equation for a fully reversible electrochemical processes, defined as: $I_{p,f}^{rev} = \pm 0.446 nFA_{real}C \sqrt{\frac{nFDv}{RT}}$ [4]. This equation directly correlates the recorded current (I) with the number of electrons in the

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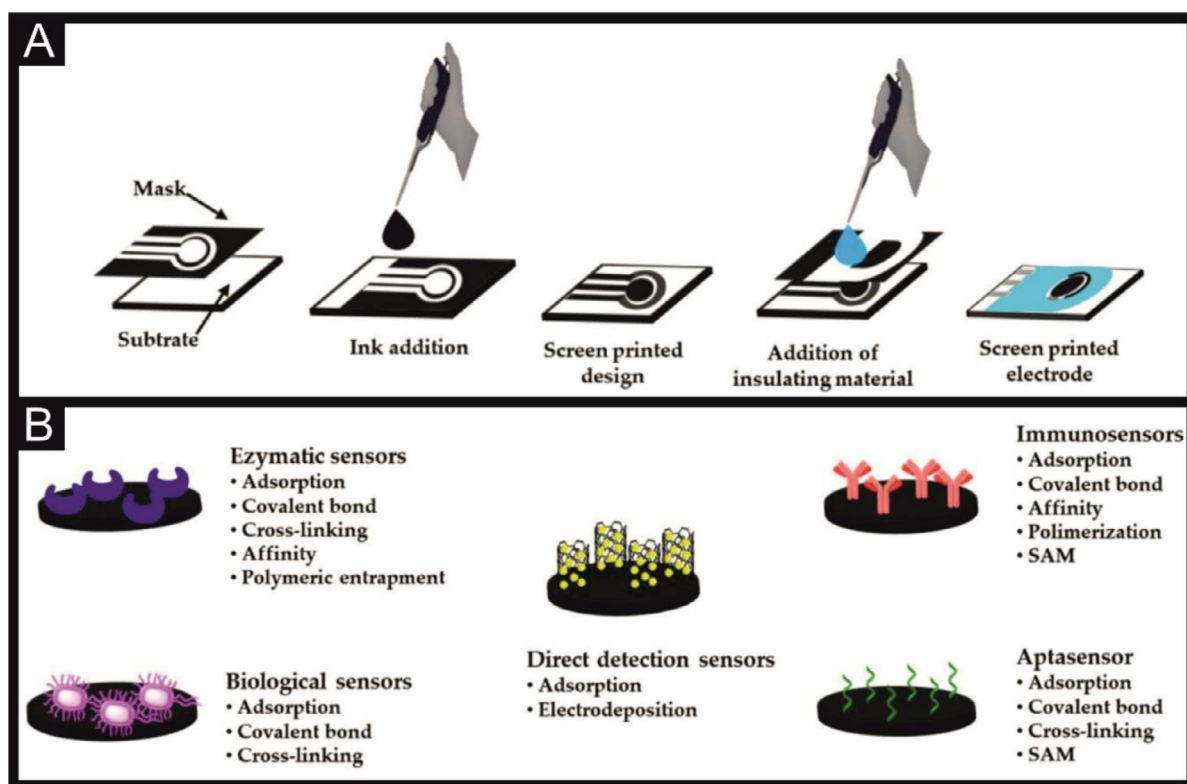


Fig. 1. Schematic representation of the manufacturing process of SPEs (A) and the different modification methods for bio-receptor immobilisation (B). Adapted and reprinted with permission from [40].

electrochemical reaction (n), Faraday constant (F), voltammetric scan rate (v), universal gas constant (R), temperature (T , in Kelvin), electroactive area of the electrode (A_{real}) and the diffusion coefficient (D). This equation, under standard conditions, becomes: $I_{p,f}^{\text{rev}} = \pm 2.69 \times 10^5 n^{3/2} A_{\text{real}} C \sqrt{Dv}$ [2]. In an electroanalytical experiment, the number of electrons participating within the electrochemical process and the diffusion coefficient will be known, the electrode area will be fixed and upon an electroanalytical measurement, the scan rate will also be selected at a chosen value. An electroanalytical experiment is then performed and as with other analytical protocols, the signal output, in regards to achievable currents, against increasing concentrations of the analytical target is explored to determine linear ranges, sensitivities, limits of detection etc. What is readily evident from the above equations is that the current is directly proportional to the analyte's concentration, effectively multiplied by 2.69×10^5 ; this amplification gives rise to the observed increased sensitivity.

Within the field of electroanalysis, voltammetric and amperometric methods are the most utilised techniques for the sensing of a range of analytes encountered in quality management [5], environmental control [6–8], forensics [9–11], biomedical [12, 13] and food [14, 15] applications, to name just a few. To name a relevant recent example, healthcare systems all over the world are in search of periodical or continuous sensor for relevant biomarkers and other analytes; there is a global attention to sensing technologies that can allow health monitoring and early diagnostics to decrease hospitalization times, hospital expenses and bills and can decrease the load of medical care and improve the overall quality of life of patients [16–18]. It is important to also note that in some particular applications, such as samples in complex matrices, the combination of electrochemistry with chromatographic, spectrophotometric and other techniques is considered a robust solution to overcome each technique's limitations and it is widely reported for electroanalytical applications [19–21].

1.2. Miniaturised equipment

Generally speaking, electrochemical methods need a sample to be analysed, a set of electrodes (WE, RE and CE), their respective connectors, a potentiostat to control the electric parameters and a computer/similar device to analyse the data [18]. Recent advances in manufacturing and computing power have allowed the miniaturization of each of these factors, pushing the electrochemistry field to a miniaturisation path. Desktop computers and potentiostat are now being substituted by handheld wireless all-in-one devices of the size of a credit/debit card, that are affordable enough to be applied to point-of-care (POC) analysis [22–27].

The analytical performance of electrochemical sensors is fundamentally linked to the material of which the WE is made of, and is one of the biggest challenges that experimentalists face when developing (bio)sensors [28–30]. Electron transport, surface-to-volume ratio, potential windows, background current and (chemical and electrochemical) stability are only *some* of the factors that affect the electroanalytical output of a given system [31, 32]. Graphitic materials (such as graphite, glassy carbon (GC) and nano-graphite) offer outstanding conductivity, chemical and electrochemical stability, versatility, wide potential windows and rich surface chemistry [33, 34]. The electrochemical performance of a given electrode can also be enhanced by modifying its surface using coatings, deposited and/or bonded materials onto the working electrode's surface [35, 36]. Coating methods such as drop-casting, spin- and dip-coating are based upon covering the surface with a solvent-modifier mix [35, 37–39]. Fig. 1 shows the manufacturing process of screen-printed electrodes (A) and the different modification methods for bio-receptor immobilisation as summarised by Pérez-Fernández *et al.* [40].

Following the general miniaturisation trend within science, electrochemistry has also moved away from 'large' instruments and materials to

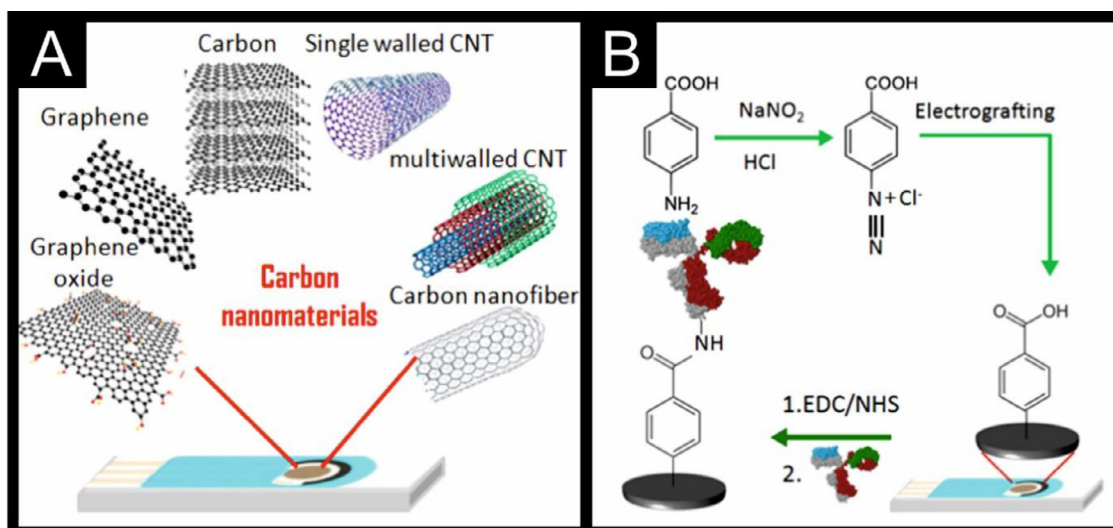


Fig. 2. A) 2D Carbon nanomaterials-modified SPEs, B) Diazonium salt-SPE functionalisation for immunosensors fabrication. Reproduced with permission from [91].

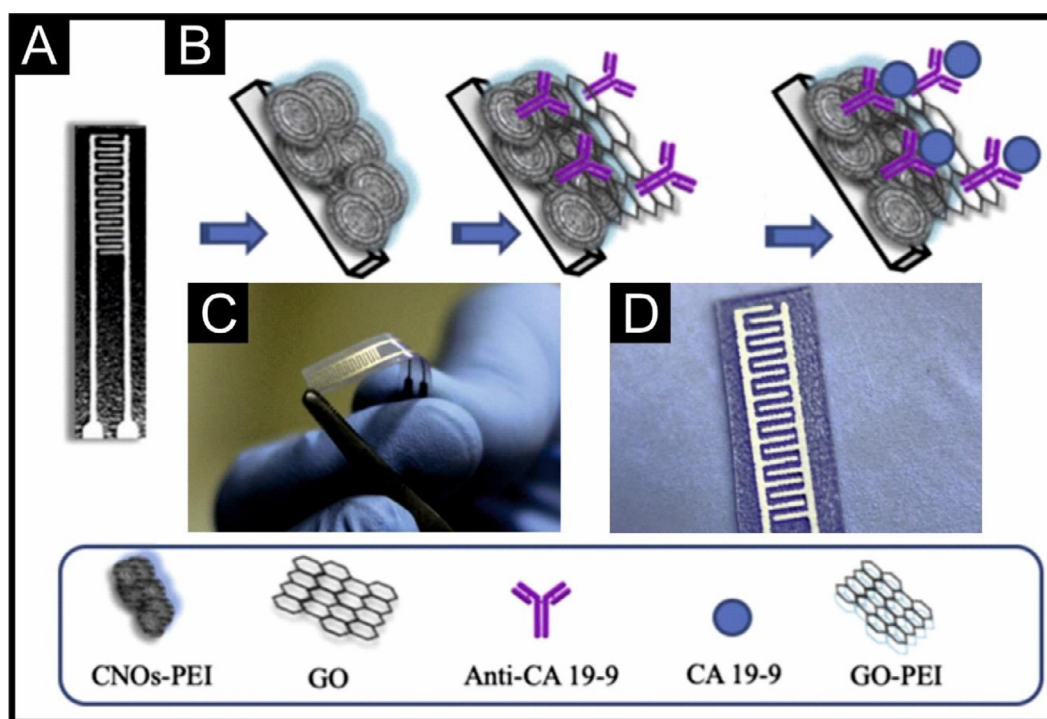


Fig. 3. Schematic illustration of a SPIDE/CNO-Go-Ab sensor screen (A), manufacturing method (B) and optical images (C and D). Adapted and reprinted with permission from [93].

simple and low-cost electrode platforms such as paste or ink deposition in a wide variety of substrates [41]. Screen-printed electrodes (SPEs) are manufactured using well-known industrial printers by depositing a combination of layers onto a flat substrate (see Fig. 1A). Screen-printing offers versatility in terms of electrode design, material compatibility and modifications yet offering mass-producible, affordable and highly reproducible sensors [42]. Current commercial laboratory tests are often complex and expensive, making them unusable for *in-situ* and point-of-care solutions for quality control, environmental and healthcare monitoring. In order to achieve and comply with quicker test turnarounds and better traceability of biosensors, a new generation of miniaturised biosensing devices is needed to be applied to the current analytical methodologies. Electrochemical techniques in combination with SPEs have been proven as capable sensors to accelerate the change from conventional

benchtop techniques/equipment to low-cost, robust and quick sensing devices.

2. Screen-printed electrodes applied towards analytical applications

2.1. Food and drinks

Although huge progress has been made in food analysis to ensure its safety and quality, it is still a paramount topic in today's everyday life. While food analysis is a complex area due to its need of multiple centralised labs (each of them specialised in different contaminants), disposable point-of-use sensors could help enhance testing methods. Food electrochemistry generally requires a liquid sample/liquid extract and

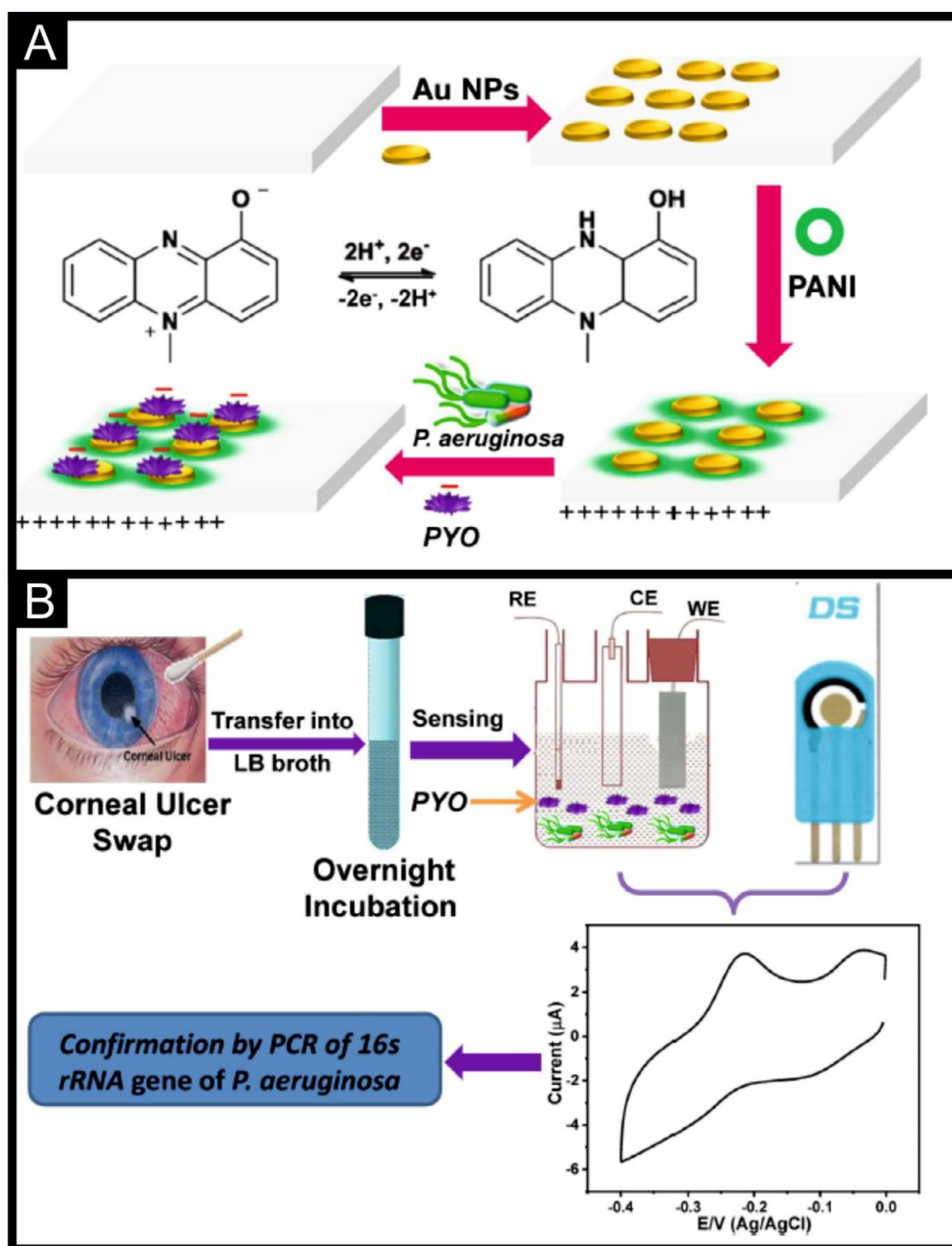


Fig. 4. (A) Schematic diagram of the PANI/AuNPs/ITO SPE fabrication and the PANI-PYO interaction. (B) Schematic representation of the electrochemical sensing of *P. aeruginosa* using the SPE sensors and an example of its electrochemical signal. Adapted and reprinted with permission from [106].

if the food is solid, pre-treatment/processing is required; generally this is why electrochemical techniques are often applied to the monitoring of drinks.

Electrochemical methods have been applied to wine analysis, from testing their alcoholic fermentation to their ageing, allergenic protein presence and sulphites content [43]. For example, Albanese *et al.* [44] showed similar limits of detection (LODs) of glucose in real red and white wines, pineapple and pear juices and dry grapes when comparing the use of Prussian blue-modified SPEs and HPLC methods [44]. Through the use of a Nafion barrier and Prussian blue, their sensor was able to have anti-interference effects against negatively charged molecules (such as the common interfering ascorbic acid), exhibiting a

highly capable performance of this oxidase-based biosensor in real food analysis. Andrei *et al.* [45] reported the determination of the antioxidant gallium oxide (GA), vitamin C, caffeic acid and quercetin when using surface modified cerium oxide-SPEs in combination with an amperometric method. The authors demonstrated their SPE sensor to exhibit lower antioxidant LOD and anti-interference behaviour than the standard TEAC (Trolox equivalent antioxidant capacity) methodology; the use of cerium-oxide based SPEs allows the detection of oxidizable phenolics in wine samples in a one-step, robust, sensitive, rapid and affordable solution without the use of enzymes [45]. Recently, Titoiu *et al.* [46] reported a label-free sensor for the allergen lysozyme in wines using AuNP-Au-SPEs (the combination of gold nanoparticles (AuNPs) on a

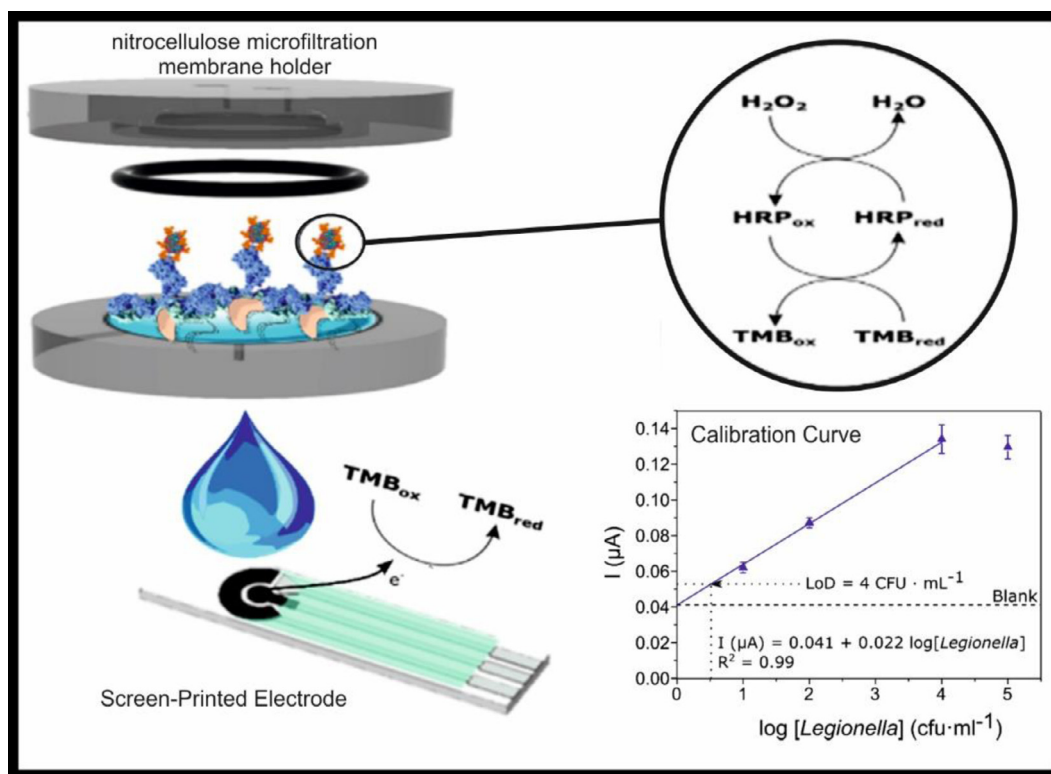


Fig. 5. Schematic diagram of the microfiltration, immunoassay, screen-printed electrode platform and calibration curve for the determination of *Legionella pneumophila* in drinking water. Adapted and reprinted with permission from [109].

gold SPE (AuSPE) is used to increase the electroactive area and facilitate the immobilization of the aptamer), exhibiting similar LODs than HPLC and colorimetric methods, highlighting the importance for strict monitoring of allergen presence during wine manufacturing; their screen-printed based aptasensor offered similar detection limits with simpler equipment, multiple use, faster turnarounds and re-usability when compared to that of HPLC [46].

In terms of food analysis, Pierini *et al.* [47] reported a novel methodology for the determination of the flavonoid taxifolin in peanut oil samples using unmodified graphitic SPEs (GSPEs) as an original, low-cost, portable and reliable method of quality control monitoring of the protected designation of origin of Argentinian Cordoba peanuts [47]. The unmodified GSPEs platform offered a low reagent consumption, no pre-treatment of the electrode needed, and offers the possibility of decentralised and disposable analysis, exhibiting a LOD of 0.021 μM, compared to LODs in the region of 0.66–0.76 μM for HPLC coupled with different detection system (ultraviolet–visible spectroscopy (UV–VIS), photodiode array detector or capillary electrophoresis) [47]. Furthermore Torre *et al.* [48] thoroughly compiled an extensive review of the use of SPEs towards bacteria and biogenic amine detection towards food spoilage control, with special attention to the suitability of SPE-based biosensors for fast decentralised on-site screening solutions. Lastly, Soulis *et al.* [49] explored the immobilisation of AChE (acetylcholinesterase) on a carbon black (CB) modified-SPE to enhance the electrocatalytic activity towards organophosphorus and carbamate pesticide detection in olive oil with LODs below the allowed limit (10 ppb) in real olive oil samples [49].

2.2. Environmental monitoring

Industrial and anthropogenic activities have increased the presence of pollutants that nowadays, can be found in air, soil, water and animal bodies. Although pollution is often directly related to more indus-

trial countries, it does not only affect developing nations, as an example many European cities *do not* meet the requirements for their air quality regulations [50]. For instance, traditional air pollutants analysers are large, heavy and expensive, with prices ranging between €5000 and €30,000 per unit [51]. While disposable sensors are not often used for gas sensing, low-cost disposable sensors *are* applied for water and soil contaminant monitoring [52]. An excellent low-cost alternative for *in-situ* environmental monitoring is electroanalysis and SPEs [53]. Traditional methods for pollutants and other environmental monitoring exhibit low LODs (*ca.* parts per billion, ppb), are highly sensitive, specific and reproducible, although they do need centralised and expensive laboratories, with sample preparation, high power consumption and require highly qualified experimentalists [54].

Inorganic, organic and biological are the three main categories of water and soil contaminants. Soilborne contaminant monitoring often requires extraction and pre-treatment of the samples, water samples often do not need any of these though heavy metal ions, pesticides/herbicides and phenolic compounds are all of high environmental concern due to their harmful effects to human health [40, 55]. García-Miranda *et al.* [8] have recently published a comprehensive review on the application of SPEs towards heavy metal ions such as Hg²⁺, Pb²⁺, Cu²⁺ and Cd²⁺ monitoring in water, highlighting their importance in the transition from lab-based to *in-situ* low-cost biosensors. Bulk modification of an SPE's ink allows a mass-production approach that offers enhanced stability and homogeneous distribution of the modifications. Bismuth films [6, 7, 56], silver [57] and gold [19, 58, 59] nanoparticles modifications have been applied to SPEs towards Cd²⁺, Pb²⁺, Cu²⁺ and Hg²⁺ sensing to name a few. An electrochemical screening assay for the determination of carbamate pesticides (carbofuran, isoprocureb, carbaryl and fenobucarb) in grain samples was demonstrated by Della Pelle *et al.* [60] where their SPEs were modified by drop-casting carbon black (CB) to increase the sensitivity, selectivity and to reduce fouling of the sensor. The carbon black-SPE (CB-SPE) sensor exhibited LODs of 80 nM for

all analytes studied, exhibiting excellent accuracy (7.8 to 9% difference) with those obtained at a UHPLC-MS/MS (ultra-high performance liquid chromatography tandem mass spectrometry). Last of note, Govindasamy *et al.* [61] reported the use of the rich edge chemistry and enhanced active sites of graphene oxide (GO) nanoribbons (NRs) to modify SPEs (GONR-SPEs) for the determination of the pesticide methyl parathion in broccoli, beetroot, tomato and Ugli fruits, showcasing excellent stability, repeatability, reproducibility and high selectivity with an LOD of 0.5 nM [61].

2.3. Forensic electrochemistry

Drugs of abuse such as cocaine, MDMA and other new psychoactive substances (NPS) such as synthetic cannabinoids, synthetic cathinones and piperazines are synthetic designer drugs that mimic their controlled substance counterparts that have been extensively explored with mass spectrometry, Raman and NIR (near-IR) spectroscopy, although more recently also with SPE sensing devices [62]. The European Monitoring Centre for Drugs and Drug Addiction (EMCDDA) has recently reported an increase in the use of such substances in Europe, showing cathinones as the leading NPS in a number of seizures, reflecting the presence of more than 560 different substances that are being monitored [63], although there are more than 10,000 different drugs can be bought on the dark web [64]. Law enforcement and health authorities worldwide are continuously challenged by the aforementioned unknown substances, with the necessity of having methods of toxicological screening of samples to identify and/or detect/quantify the new compounds entering the market [65, 66].

Most common screening methods are quick antibody-based colorimetric kits, which offer only qualitative results [67]. Portable electrochemical sensors based on GSPEs enable police forces to perform vital in-situ monitoring and determination of prohibited compounds, taking advantage of their portability and excellent performance to develop into-the-field sensors able to detect NPS quickly, in a variety of sample matrices and within a wide concentration range [20, 68, 69]. Unmodified graphitic SPEs (GSPEs) were shown as mephedrone metabolite sensors in human urine by Elbardisy *et al.* [69], exhibiting LODs of 6.34 and 3.87 $\mu\text{g mL}^{-1}$ for 4-MC and 4-MMC-R respectively, with no interference effects of paracetamol, 1-methyluric acid and 4-acetamidobenzoic acid being observed when detecting 4-MC [69]. Lima *et al.* [70] explored the determination of MDPV (3,4-methylenedioxypyrovaleron) cathinone with a GSPE, reporting a LOD of 0.5 μM and no interference effects from caffeine, paracetamol or glucose. Bare GSPEs were also reported for the electrochemical determination of fentanyl, exhibiting no interference with methamphetamine, caffeine nor acetaminophen by Ott *et al.* [71], with special interest to their oral fluid application for driving under the influence of drugs (DUID) situations. Unmodified GSPEs have also been recently reported towards the determination of MDMA, exhibiting a LOD of 1.83 μM and being proposed as an alternative *in-situ* sensing method [72].

Other forensic applications explored by portable electrochemical systems include commonly misused prescription medications, such as rohypnol and atropine. Smith *et al.* [73] first reported the use of bare GSPEs for the determination of rohypnol in both Coca ColaTM and alcopop WKDTM drinks without sample preparation, exhibiting an LOD of 0.47 $\mu\text{g mL}^{-1}$ [73]. Later, Tseliou *et al.* [74] applied an enzymatic electrochemical method with GOx/GluHD-modified FeGSPE (glucose oxidase/glucose hydrogel modified iron-sparked graphite SPEs) in a wider range of untreated and undiluted spiked samples in a range on non- and alcoholic spiked samples with alcohol content up to 40% v/v [74]. Also the determination of atropine in spiked diet Coca ColaTM using GSPE, exhibiting a LOD of 18.4 μM , without interference effects from caffeine nor ascorbic acid has been reported by Ramdani *et al.* [75]. Lastly, Ping *et al.* [76] designed a novel bulk modification for dopamine determination in body fluids using graphite, cellulose acetate and ionic liquid OPPF (n-octylpyridinium hexafluorophosphate). The use of the

ionic liquid enhanced its biocompatibility and the graphite-cellulose acetate modification decreased the overpotentials of ascorbic and uric acid oxidations due to the fast electron transfer rate, allowing the determination of dopamine in pharmaceutical preparation without interferences, reporting an LOD of 0.5 μM [76].

2.4. Cancer biomarkers

Cancer is one of the more prevalent diseases in the 21st century worldwide. Screening, early diagnosis, monitoring and treatments with non-invasive and robust methods for cancer patients are at the forefront of the general scientific priority in order to increase both expectancy and quality of life [77]. Biopsies and imaging techniques are the current methods for cancer diagnosis, that need an already existent tumour to be detected and require highly trained technicians and physicians in addition to costly equipment to understand their results [78]. Early diagnostics try to tackle the undetected spread of cancer cells by analysing blood, urine and saliva samples with minimum or non-invasive assays and techniques. Abnormal cell division, or cancer progression, can be tracked and followed by specific biomarkers that otherwise would not be found (at all or in high quantities) [79]. Protein biomarkers allow the predictive diagnostics, recurrence and staging of a cancer, providing a patient's health snapshot [80]. Current commercial kits such as ELISA (enzyme-linked immunosorbent assay), LC-MS (liquid chromatography-mass spectrometry), Luminex and single-molecule counting (Simoa-HD) have trouble detecting low concentrated biomarkers in complex matrix with a high concentration of other proteins within the sample [81–83]. It is because of this, that electrochemical biosensors are now considered for cancer diagnostics due to their ultra-sensitivity and selectivity, low-cost, fast turnaround yet large scale production, miniaturization and multiplexing capabilities [84–87]. The integration of electrochemical biosensors for “cancer-on-a-chip” detection methods is a growing research trend, where the electrochemical targets are at the molecular (DNA, RNA and proteins), organelle (exosomes) and cell levels (phenotypic and metabolism analysis, drug sensitivity monitoring and cell counting) [88]. See the thorough reviews by Rusling *et al.* [83] and Feiyun *et al.* [88] for further electrochemical cancer biomarkers literature knowledge and trends.

The development of affinity biosensors requires a suitable modification and immobilisation design for the overall performance of the electrode. These can be physical or chemical strategies, based on electrostatic interaction or covalent coupling between the biomolecule and the surface respectively. Currently diazonium electro-grafting, depicted in Fig. 2, is one of the most used functionalisation techniques due to its simplicity and versatility to provide the attachment of an organic later to the surface of the working electrode [89–91]. Yáñez-Sedeño *et al.* [89] published a comprehensive review on diazonium salt-modified SPE biosensing platforms, paying special attention to the importance of coupling multiplexed SPEs with the electro-grafting method to individually functionalise each working electrode, increasing the possibilities of mass-production of integrated electroanalytical platforms for simultaneous determination of biomarkers [89]. Corrigan *et al.* [92] recently reported the use of a multiplexed commercial eight GSPE working electrodes (2.95 mm in diameter with a concentric common carbon counter electrode and a Ag/AgCl reference electrode placed in the centre of the circle) for the quantification of mutant genetic sequences from clinical samples, showcasing high sensitivity and specificity for their target sequences. The electrode material was chosen to be carbon due to its well establish surface functionalisation chemistries and anti-biofouling properties; a diazonium compound, followed by NHS-EDC (N-hydroxysuccinimide-1-ethyl-3-(3-dimethylaminopropyl) carbodiimide) coupling of amine-tagged DNA was the surface modification [92].

One of the most widely used early biomarkers for pancreatic cancer is CA19-9 (carbohydrate antigen 19-9), although current methodologies have low sensitivities. To showcase a different approach, as shown in

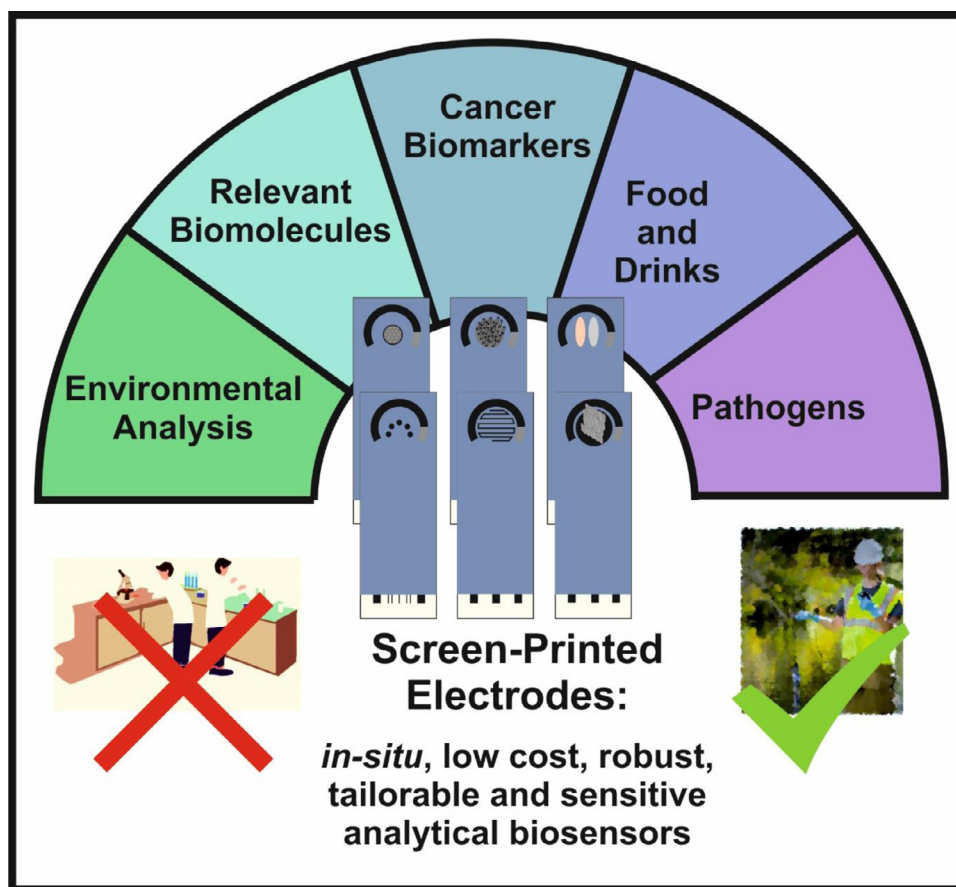


Fig. 6. Schematic representation of possible applications for tailored, affordable and *in-situ* screen-printed electrochemical sensing platforms.

Fig. 3. Ibáñez-Redín *et al.* [93] recently reported capacitive biosensors using carbon nano-onions (CNOs) and graphene oxide (GO) low-cost interdigitated screen-printed electrode (SPIDE) for the immobilisation of anti-CA19-9 antibodies, exhibiting a LOD of 0.12 U mL^{-1} , which is within the relevant range for early pancreatic cancer detection. Moreira *et al.* [94] reported the use of an SPE platform for the low-cost point-of-care screening of the carcinogenic embryonic antigen (CEA). Their proposed biosensor uses a plastic antibody entrapped on polypyrrole (PPy), for the later elimination of the antibody which will house the selective adsorption of CEA to MPPy (PPy after CEA removal). The reported approach displayed simple design, short measuring times, precise reading of <5% deviation and LODs in the order of $\sim 1 \text{ pg mL}^{-1}$ [94].

Note that it has been reported that immunoassays have the downside of reliability of immobilisation techniques, orientation of the transducer, quality of antibodies and limited shelf life [95]. Molecularly Imprinted Polymers (MIPs), or synthetic antibodies, offer biological recognition elements with superior stability, tailorability and cheaper production [96]; see reference [97] for further information.

2.5. Pathogen electroanalysis

Microorganisms such as bacteria, fungi, protozoans and other infectious agents including viruses and prions that are harmful are called pathogens. As an example, in 2020 there was a global pandemic caused by the COVID-19 disease (SARS-CoV-2 virus) for which rapid and sensitive detection methods were essential in the quick diagnosis of patients, which in turn lowered the infection and mortality rate. Within the last few decades conventional approaches such as conventional culturing, straining, molecular methods (such as ELISA and PCRs (polymerase chain reaction)), microscopy-based and mass-spectrometry techniques have been applied to the identification and quantification of

pathogenic agents [98]. Immunoassays and DNA-based assays, such as ELISA and PCR are the most common identification techniques for these pathogens. Immunoassays can directly and indirectly detect the presence of pathogens by using bio-recognition and target elements that target pathogen epitopes and generated antibodies respectively. DNA-based techniques are applied to cases in which there is a reduced/lack of antibody availability, targeting antibodies, genes responsible of toxin production, nucleic acids, viruses, cells and toxins themselves. Antibodies, aptamers and imprinted polymers are the most common used biorecognition elements [18, 99]. Electrochemical biosensors towards the detection of pathogens are a growing topic in today's literature [100, 101]. Electrochemical pathogen biosensors allow the *in-situ* analysis of complex matrices with little or no sample preparation using the working electrode as a transducer and an immobilised recognition probe/molecular probe. For further information regarding pathogen detection using electrochemical methods readers are directed to references [101–105]. As stated by Simoska [98], further efforts in electrochemical sensors for pathogens are significantly needed in order to shorten the analysis time, reduce or eliminate sample preparation, overcome bio-fouling effects and their performance in true complex media and clinical samples.

Fig. 4 shows the electrochemical detection of *Pseudomonas aeruginosa* using an ultrasensitive SPE modified with polyaniline (PANI)/gold nanoparticle (AuNPs)/iridium tin oxide (ITO) via the detection of pyocyanin (PYO) in real corneal ulcers samples was reported by Khalifa *et al.*, exhibiting a 100% agreement with the classic molecular method in sensitivity, specificity, positive and negative predictive values when comparing the SPE, conventional and automated methods (phenotypic and PCR methods) [106]. Radi *et al.* described the electrochemical behaviour of a mycotoxin from the *Fusarium* fungi that infects cereal crops, Zearalenone (ZEA), reporting the quantitative analysis of ZEA

in cornflake samples using commercial bulk-modified single-walled carbon nanotube screen-printed electrode (SWCNT-SPE). An LOD of $2.5 \mu\text{g L}^{-1}$ using in real samples, which is *ca.* 10 times less than the maximum level allowed of ZEA in food by the European Union (EU) was reported [107]. Another example of microbial biosensor has been reported by Uria *et al.* [108], in which they propose the immobilisation of bacteria on SPE surfaces by using trapping them in a cellulose matrix. Following that, they tested toxicity using a lateral flow biosensing platform for the rapid monitoring of toxic compounds with an electrode polarization in the presence of ferricyanide and glucose. This platform was reported as a cost-effective and easily manufactured with excellent shelf life for up one year towards the detection of bacteria-based toxicity [108]. Recent work from the same researchers, as shown in Fig. 5, cleverly reported an electrochemical immunoassay system that reduces the detection time from 10 days (conventional culture-based methods) to that of only 2–3 hrs for the detection of *Legionella pneumophila* in drinking water [109]. Their methodology is based on a microfiltration membrane that acts as support for both sample concentration and antigen-antibody reaction (horseradish peroxidase (HRP) enzyme and 3,3',5,5'-Tetramethylbenzidine (TMB) mediator), for a chronoamperometric measurement using GSPEs, with the additional benefit of using a single antibody system which lowers the cost for each test [109].

To summarise, Fig. 6 highlights the different topics to which screen-printed sensor platforms can be applied to, including a representation of the different shape designs from single-, dual- to multiplexed working electrode configurations and 2D modifications materials. This short literature review discusses and showcases the work of many researchers with their successful applications of SPEs to a plethora of analytes and analytical chemistry areas, demonstrating that SPE platforms offer a reliable, robust, sensitive and totally tailorable approach as an alternative to traditional laboratory standard techniques. However, improvements in bringing these solutions to market to truly speed up test turnarounds, improve early detection and achieve mass environmental monitoring are still needed.

3. Conclusions

Electrochemical biosensors, when coupled with screen-printed electrodes, provide an alternative to analytical techniques for in-to-the-field screening and monitoring solutions in a plethora of fields such as food, environmental, forensics and cancer biomarker analysis. This short review shows that electrochemical SPE sensors provide sensitive, rapid and affordable sensors, that due to progress in nanomaterials and biotechnology advances offer a powerful platform for recognition elements to be used *in-situ* monitoring biosensors. With the introduction of stricter pollutant limits and more sensitive biomarker diagnostics requirements, there is an urgent need for transition from traditional lab-based techniques to miniaturised, cheaper and quicker tests for health and environmental monitoring; from the above summary, it is likely that SPEs can help bridge this gap. However, further research still needs to be undertaken to address challenges such as selective multi-analyte determination, be able to perform reliability within complex matrices and more work needs to be independently validated against gold standard laboratory based instrumentation. Other areas that SPEs can provide significant benefits, with further research, are screen-printed based wearable sensors and/or *in-situ* healthcare applications etc. This is a unique opportunity for SPEs to become a key player in the low-cost sensor market, continuing to build upon past- successes, *i.e.* the glucose sensor.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

Funding from Innovate UK (KTP Reference: 11606) is acknowledged.

Additional information

The authors declare no competing financial and/or non-financial interests in relation to the work described herein.

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